ONR Contract N00014-87-C-0339: October 1986-October 1991

Final report

Investigations of the Epitaxial Growth and Characterization of High Perfection Magnetic Thin Films.

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Final Report for ONR Contract N0001-87-C-0339.

Investigations of the Epitaxial Growth and Characterization of High-Perfection Magnetic Thin Films.

ABSTRACT

Methods of preparing high-perfection single crystalline films of 3d elements (including Fe, Co) and magnetic rare earths on semiconducting GaAs substrates have been investigated. The use of seed layers of Fe or Co (a few monolayers thick), deposited onto GaAs(001), followed by a Ag(001) film, provided a suitable template for Fe epitaxy. This method avoided interfacial chemical reactions between Fe and the semiconductor, at the expense of introducing coherency strain and tilted epitaxy of the Fe. In the case of rare earths, intermediate seed films of rare earth trifluorides, grown onto GaAs($\overline{111}$) provided a template for growth of c-axis oriented rare earth films and sandwich structures. These methods have enabled the effects of coherency strain on magnetic properties of Fe and rare earths to be investigated.

Using an automated X-ray photoelectron diffraction (XPD) system we have investigated the growth and interfacial mixing of several key epitaxial systems including Co/Pt, Co/Ag, NdF_3/LaF_3 and Pt/GaAs. Interfacial mixing in the interfaces of Co/Pt superlattices was confirmed, resulting in an alloyed region containing the ordered $L1_2$ phase: $CoPt_3$. Spin-polarized photoelectron diffraction (SPPD) experiments on MnF_2 suggest a new , high-temperature magnetic ordering transition at 380K which is 313K above the Néel temperature.

1. Introduction.

The overall objective of this contract was to develop ways of preparing high perfection single crystalline films of 3d elements (including Fe, Co) and magnetic rare earths on semiconducting GaAs substrates. To accomplish this, it was necessary to introduce (for the first time in the field of magnetic thin films) cross section transmission electron microscope techniques and high resolution X-ray diffraction techniques to study the microstructure and perfection of the metal-semiconductor interfaces. New seed film techniques were developed, allowing the effect of coherency strain on magnetic properties to be investigated. The work proved to be influential in that the seeding techniques were valuable in controlling the orientation of magnetic superlattices (Co/Pt) on GaAs and were adopted by other groups in preparing the Fe/Cr (001) oriented superlattices on GaAs(001) in which giant magnetoresistance was discovered.

During the 2 year extension of this contract (from October 1989 to October 91) emphasis was placed on the use of XPD and SPPD to investigate several key magnetic systems. The power of these technique was well demonstrated.

This report summarizes the objectives, results achieved and a list of publications resulting directly from the contract. It contains copies of the year-end reports submitted as part of the reporting process.

2. Objectives.

This contract had several key objectives:

(a) Growth of Fe films on GaAs and In_xGa_{1-x} to achieve lattice-matching across the metal-semiconductor interface.

(b) The use of cross-section TEM techniques to examine the microstructure and strain relaxation of epitaxial Fe/GaAs films.

(c) A comparison between the observed magnetic and structural properties of epitaxial Fe films on semiconductor substrates with the predictions of a parallel ONR

contract with IBM (Principal Investigator, F. Herman, ONR contract N00014-85-C-0467).

(d) The growth of rare earth metal films and superlattices on suitably lattice matched substrates.

During the extension period, the main objective was to use XPD and SPPD to probe key but poorly understood magnetic systems. The results for XPD of Co/Pt interfaces and for SPPD of MnF_2 are summarized.

2. Experimental Description.

In all the studies described in this report, films were deposited in a molecular beam epitaxy system (VG80M supplied by VG Semicon, UK). The sources for the magnetic elements Fe, Co, rare earth metals Dy, Ho, Er, Y and Pt, were electron gun evaporation sources (Temescal). For Ag and Mn, effusion sources were used. The growth chamber pressure prior to film growth was $<1.10^{-10}$ mB. During film growth this pressure increased to 1 to 4.10^{-10} mB. The semiconductor substrates in this work fall into two different categories. For studies of Fe epitaxy, GaAs (001) oriented wafers, onto which had been grown undoped GaAs or $In_xGa_{1-x}As$ epilayers, were used. These layers were grown in a separate MBE machine, dedicated to high purity III-V semiconductor film growth. The epilayer surface was capped with an amorphous film of arsenic (deposited from an As_2 source onto a cold (<0°C) substrate. After a brief exposure to air the substrate was transferred to Almaden Research Center in a vacuum flask and loaded into the MBE machine. The arsenic cap was desorbed immediately prior to magnetic film growth by heating the substrate to about 450C for a few minutes. In all cases the sample surface exhibited a (1x1) reconstruction and an Auger spectrum indicated only about 1-3% ML carbon was present as an impurity.

For epitaxial growth of rare earth films and sandwiches, GaAs($\overline{111}$) oriented substrates were used. Epitaxial rare earth trifluoride films were used as seed films for the rare earths. The hexagonal-structure fluorides grew with the c-axis along the film normal and the c-axis of the rare earths followed this orientation.

A variety of analytical techniques were used for structural and magnetic characterization of the films. In situ structural techniques included, auger electron spectroscopy, X-ray photoelectron spectroscopy, reflection high energy electron diffraction (RHEED) and low energy electron diffraction (LEED). X-ray diffraction, including both in-house double crystal diffraction and synchrotron X-ray diffraction techniques were used. Cross section TEM was carried out in-house and at Stanford University in collaboration with an IBM-funded research student (C.J. Chien). Magnetic characterization techniques included, ferromagnetic resonance, SQUID and vibrating sample magnetometry.

3(a) Epitaxial Growth of Fe films on GaAs and InxGa1-xAs

Epitaxial growth of Fe films directly onto GaAs and onto lattice matched $In_{0.2}Ga_{0.8}As$ substrates (without buffer layers of Ag), at substrate temperatures of 20 or 180°C was studied. The results are described in detail in several publications [1-4]. The key results were:

1. Fe films grew in a parallel epitaxial orientation with: Fe[001]//GaAs[001] and Fe[100]//GaAs[100], as expected from the small misfit (about 1.4%) between the doubled Fe unit cell (2a=5.732Å, 25°C) and the unit cell of GaAs (5.6537Å, 25°C) [1].

2. The growth mode of the Fe was by island growth and coalescence at 20 and 180C [1].

3. For both GaAs and $In_xGa_{1-x}As$ substrates there was an interfacial reaction indicated by the presence of As (and In in the case of $In_xGa_{1-x}As$) on the top surface of even 50ÅA thick Fe films [1].

4. Misfit between Fe and GaAs was partially accommodated by misfit dislocations in the GaAs which were imaged by cross-section TEM[2]. The dislocation density in Fe films (300-900A-thick) was similar for growth on GaAs and InGaAs substrates despite lattice matching in the latter case. This may be due to penetration of dislocations from the GaAs/InGaAs interface into the Fe film and partially to the tendency of GaAs to take up some of the misfit between Fe and GaAs in the former structures. Quite sophisticated blocking schemes, designed to block dislocation propagation into the top of the $In_{0.2}Ga_{0.8}As$ film, were only partially successful. These schemes included[1] insertion of a strained layer superlattice (4.5 periods of 100Å $In_{0.4}Ga_{0.6}$/ 100Å GaAs) within the $In_{0.2}Ga_{0.8}As$ and compositionally grading from the GaAs to the $In_{0.2}Ga_{0.8}As$.

5. FMR linewidths for 300 to 900Å-thick Fe films did not differ significantly for Fe/GaAs in comparison with Fe/$In_xGa_{1-x}As$ consistent with a similar density of dislocations (10^8 to 10^9 cm^{-2}) in both cases.

6. Linewidths of Fe films grown at 180C on GaAs or InGaAs were in the range 50-70 Oe and narrower than for comparable films grown on ZnSe substrates [1] indicating that the films were of the highest structural quality yet prepared.

7. Analysis of the FMR data showed that the extremal interfaces of 300-900Å-thick Fe films were , in all cases, unequal in terms of surface/interface anisotropy energy Ks. This is probably due to a reduced magnetization region in a thin (<20A thick) region of the Fe adjacent to the Fe/GaAs interface.

In order to avoid the interfacial reaction between Fe and GaAs the insertion of an epitaxial Ag film proved highly effective. However, in order to achieve a single

sharply defined epitaxial orientation: Ag(001)// GaAs(001); Ag[100]//GaAs[110], it was necessary to insert an initial seed film of about 3-6 ML of Fe on the initial GaAs surface. Without this initial Fe seed film the Ag grew in a quite different orientation: Ag[110]//Fe[100]. This orientation of Ag was later used in growing seeded, epitaxial (110)-oriented Co/Pt superlattices (see section 6(a)). It was found that Co was also effective as a seed film for Ag(001) epitaxy. Onto this Ag (001) template, Fe grew with Fe(001)//Ag(001) and with a 45° rotation of the Fe unit cell: Fe[100]//Ag[110]. In this orientation, the Ag and Fe lattices have an in-plane misfit of only 0.8%.

3(b) Characterization of epitaxial Fe/Ag/Fe/GaAs(001) structures.

Cross section TEM images[3,5] of the Ag/Fe/GaAs(001) interface region revealed that the interfaces were largely coherent. Coherency strain in Fe overlayers, as thick as 2000Å, grown on the Ag template film was confirmed by extensive X-ray diffraction studies[4] (see paper in Appendix). These studies also revealed that both the Ag and Fe lattices were tilted significantly and that the tilts were in the same plane as the tilt (due to miscut) of the [001] axis from the substrate surface normal. This tilt is a symmetry breaking effect and may be responsible for the 2-fold magnetic anisotropy [4] which is superimposed on the 4-fold anisotropy expected for Fe(001) films.

X-ray rocking curves of Fe films near 2000Å thick exhibited rocking curve widths (for Fe(022) and Fe(002) for CuK alpha radiation) of $\simeq$800 arc sec.. This is the narrowest linewidth yet reported for an Fe film of this thickness but is still an order of magnitude larger than for the perfect crystal case. This is consistent with a high density ($>10^8$) of dislocations in the film.

4. Epitaxial Growth of Rare-Earth Metals on Lattice-Matched Films of Rare Earth Fluorides.

As in the case of transition metals, epitaxy of rare-earth metals directly onto the surface of compound semiconductors results in an interfacial reaction. We have implemented a new approach in which a hexagonal-structure rare-earth fluoride film is interposed between the GaAs and the rare earth metal. The rare-earth fluorides vaporize as monomer molecular units and grow by a simple process of sublimation. Moreover, we have shown[6,7] that the light rare earth fluorides (for example, LaF_3, NdF_3) grow epitaxially on GaAs $(\overline{1}\overline{1}\overline{1})$ surfaces forming a very flat overlayer, tending to planarize roughness in the underlying GaAs surface. A variety of rare-earth sandwich structures were grown using this seeding approach with the aim of probing the effect of strain on the magnetic properties of rare-earths, especially Dy.

In the first series of experiments, LaF_3/Dy/LaF_3/GaAs $(\overline{1}\overline{1}\overline{1})$ sandwiches were prepared with several Dy thicknesses. Magnetisation of these structures, as a function of temperature, was measured using a SQUID. The results show[4] that for very thin (75Å) Dy films the temperature of the transition from the helical (antiferromagnetic) state to the ferromagnetic state was similar to that in thick (2888Å) films. This result is in sharp contrast to that seen for Y/Dy/Y sandwich structures grown on sapphire basal plane substrates [8], where the ferromagnetic transition was completely quenched for thin (<100Å thick) Dy films. In the Y/Dy/Y structures, the large (1.6%) misfit between Y and Dy results in lattice clamping of the Dy to a tensile state inhibiting the ferromagnetic ordering transition. However, the Dy/LaF_3 interface is either incommensurate or discommensurate and lattice strain is not transmitted into the Dy film [10] resulting in negligible strain effects on the ferromagnetic ordering transition. The absence of lattice strain in Er films down to 50Å and in Ho films down to 25Å in thickness was also confirmed[10]. The ferromagnetic ordering transition in 100Å-thick Ho films was at a similar temperature to bulk Ho and the saturation magnetization was also similar, consistent with the absence of strain in a high perfection film. This behavior of strain-free metals within

an epitaxial sandwich structure is analogous to that of epitaxial semiconducting PbTe films grown onto CaF_2 or BaF_2 films. Misfit stress, caused by thermal cycling and differential thermal expansion/contraction is taken up by dislocations formed within the fluoride rather than in the semiconductor. It is well known that dislocation mobilities in fluorides are very large, even at low temperatures. This fact is utilized in minimizing the effects of strain on narrow band gap semiconductors, such PbTe. Zogg et al [9] have shown that CaF_2 can be used as a compliant underlayer to take up misfit strain when PbTe/CaF_2 structures are repeatedly cycled between room temperature and 77K.

In the second series of experiments, a series of sandwich structures comprising LaF_3/Er/Dy/Er/LaF_3/GaAs($\overline{111}$) were prepared to study the magnetic behavior of Dy subjected to coherency strain from lattice clamping to the thick Er adjacent layers [10] (see paper in Appendix). In one such structure a 500Å-thick Dy film was sandwiched between a 2000Å-thick Er underlayer and a 1000Å-thick Er overlayer. The Er underlayer was unstrained and relaxed to its bulk lattice constant. The Dy film was in a state of lateral compression due to its smaller (by 1.1%) lattice constant. X-ray diffraction [11] confirmed that the sandwiched Dy film was indeed in a state of lateral compression and that its perpendicular expansion was $0.27 \pm 0.02\%$. This value is roughly half that expected for full coherency strain, probably due to partial accommodation of the misfit by dislocations. Nevertheless this residual strain had a dramatic effect on the magnetic ordering behavior of the Dy. The helical to ferromagnetic ordering transition was 25-30K higher than for bulk Dy. This enhancement is believed to be due to the perpendicular strain of the Dy film which causes the Dy lattice to be closer to the lattice dimensions of the ferromagnetically ordered state. In other words, the energy barrier for the helical to ferromagnetic transition is lowered. An even larger enhancement should be possible

for larger coherency-induced in-plane compressive strain in Dy. This has been demonstrated recently for Lu/Dy/Lu sandwiches[11]

The key results from this study of rare earth epitaxy on GaAs can be summarized as follows:

1. A new seed layer approach for growing magnetic rare-earths onto GaAs substrates has been demonstrated.

2. The rare earth trifluorides (LaF_3, NdF_3) form incommensurate or discommensurate interfaces to the rare earths and strain is not transmitted to the buried rare earth metal.

3. There is an interfacial reaction between the rare earths and the fluoride seed film but this is limited. For example, Ho films as thin as 100A exhibit bulk saturation magnetization indicating that any reacted component of the Ho is probably less than ~10Å thick.

4. A sandwich structure, designed to introduce in-plane compressive strain into a Dy film, and to avoid interfacial contamination of the Dy gas been demonstrated. A large (25-30K) enhancement in Dy Curie temperature has been found.

5. Contact with parallel ONR contract: N00014-85-C-0467

The main results of the above contract related to self-consistent band structure calculations (using local spin density functional theory) for lattice-matched, hcp Co/Cr superlattices. Interlayer coupling was found to depend on the number of Cr planes in the spacers, as found in experiments on bcc Fe/Cr superlattices and sandwiches by Grunberg et al[12a]. It is not clear whether epitaxial hcp Co/Cr structures can be prepared on semiconducting substrates. We have concentrated in this project on controlling the orientation of Ag seed films on GaAs(001) which provide the basis for epitaxial Fe/Ag sandwiches and superlattices. Using the same seeding technique it may be possible to grow bcc Co/Cr superlattices though the

metastable nature of bcc Co may limit the thickness of bcc Co in the superlattice. Our seeding technique permits growth of bcc Fe/Cr(001) superlattices on GaAs. Indeed, in the recent work on giant magnetoresistance in Fe/Cr superlattices[12] our use of a thin Fe(001) seed film on GaAs was utilized to seed bcc Cr(001) and to set the (001) orientation of the entire Fe/Cr(001) superlattice.

6. XPD and SPPD of key magnetic systems.

Several key magnetic systems were investigated using XPD and SPPD.

(a) XPD of Co/Pt superlattices.

Polycrystalline Co/Pt superlattices, deposited by sputtering or evaporation, exhibit a large perpendicular magnetic anisotropy and are of interest as media films for magneto-optical recording.The origin and mechanism of this anisotropy is not understood. To throw new light on this problem we used new seeding techniques, developed in our prior work under this contract, to prepare for the first time, highly-oriented epitaxial superlattices along the three major crystallographic directions: [001], [110]and &brk.111] and to investigate the orientation dependence of magnetic anisotropy [13] (see paper in Appendix). GaAs $[\overline{111}]$ and (001)-oriented substrates were used. Growth of Ag seed layers were used to seed the orientation of the Co/Pt superlattices: Ag(111)/GaAs($\overline{111}$), Ag(001)/Co(001)/GaAs(001) and Ag(110)/GaAs(001). The room temperature magnetic anisotropy of the superlattices was found to differ strongly with growth orientation (for Co 3Å - Pt 17Å superlattices) and differences in interfacial disorder were suggested by X-ray diffraction. To probe the interfaces more directly, during their formation, we used XPD [14] (see paper in Appendix). The essential results were (a) that interfacial mixing occurred for all three orientations to a similar extent: 2-4 ML at each interface and (b) that more coherency strain was evident in the case of (001) than the (111) growth orientation. We conclude that models for the perpendicular magnetic anisotropy should include interfacial

mixing and alloying as well as coherency strain. Indeed, these studies led directly to the discovery of perpendicular magnetic anisotropy in co-evaporated alloy films of $CoPt_3$ which were subsequently used in high CNR magneto-optical recording demonstrations at IBM.

(b) SPPD of MnF_2.

SPPD has been shown [15] to be a powerful probe of short-range magnetic order in antiferromagnetic insulators such as MnO and $KMnF_3$. Our initial results of SPPD of MnF_2 under this contract, show that there is a new, high-temperature, short-range magnetic ordering transition at 380K which is 313K above the Néel temperature for bulk MnF_2.

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Appendix 1.

Selected key papers on work supported by this contract.

RJ 6648 (63959) 1/19/89
Materials Science

LONG-RANGE COHERENCY STRAIN AND TILTED EPITAXY IN Ag-Fe-Ag SANDWICH STRUCTURE ON GaAs(001)

Mat. Res. Soc. Symp. Proc. 102, 483 (1988)

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Abstract:We find that epitaxial Fe films sandwiched between epitaxial Ag films grown on GaAs (001) substrates possess residual coherency strain at a thickness of 2000Å. The [001] directions of the Fe and Ag films are tilted with respect to the GaAs [001] axis. The tilts are coplanar with the tilt of the substrate surface normal to the [001] axis of GaAs and are qualitatively consistent with a recently proposed model for tilted epitaxy.

LONG-RANGE COHERENCY STRAIN AND TILTED EPITAXY IN Ag-Fe-Ag SANDWICH STRUCTURES ON GaAs(001) SUBSTRATES.

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ABSTRACT.

We find that epitaxial Fe films sandwiched between epitaxial Ag films grown on GaAs (001) substrates possess residual coherency strain at a thickness of 2000Å. The [001] directions of the Fe and Ag films are tilted with respect to the GaAs [001] axis. The tilts are coplanar with the tilt of the substrate surface normal to the [001] axis of GaAs and are qualitatively consistent with a recently proposed model for tilted epitaxy.

INTRODUCTION.

Recent studies of magnetism in epitaxial Fe/Ag structures have revealed a variety of interesting properties. For example , studies of the magnetic properties of epitaxial Fe films grown on Ag(001) surfaces [1,2] and Fe-Ag multilayers grown on Ag(001) films on GaAs(001) surfaces [3] show that the Fe has perpendicular anisotropy for thicknesses in the range 2-5 monolayers. In addition , transient paramagnetism has been detected [4] in Ag films on Fe(001) surfaces due to spin-polarization of conduction electrons crossing the Fe-Ag interface. Such structures have not been structurally characterized. In particular , the state of strain of the films and the microstructure of the various interfaces have not been examined. In this paper, we report the results of an analysis of coherency strain in epitaxial Fe-Ag sandwich structures grown by MBE on GaAs(001) surfaces. Double-crystal X-ray diffractometry was used to determine coherency strain and TEM (transmission electron microscopy) was used to probe the microstructure of the GaAs-Fe and Ag-Fe interfaces.

FILM GROWTH AND IN SITU CHARACTERIZATION

The substrates for Fe and Ag epitaxy were undoped films of homoepitaxial GaAs , grown by molecular beam epitaxy at IBM T.J. Watson Research Center. The surfaces of the substrate wafers were inclined at about 2 ° to the (001) plane , the tilt being along a <100> direction. Following GaAs epitaxy the surface of the sample was protected by an overlayer (cap) of arsenic which was deposited at ~250K. After a brief exposure to air the sample was transferred in a vacuum flask to Almaden Research Center and loaded into a VG80-M, MBE machine (VG Semicon Ltd.). The arsenic cap was desorbed by heating the sample to ~450°C for a few minutes. The sample surface exhibited either a (1x1) or (2x4) reconstruction , both of which were indicative of an As-stabilized surface. The results of this study did not depend on which of these reconstructions existed prior

to epitaxy. Auger electron spectroscopy indicated ~1-3% monolayer of carbon as an impurity. No other surface impurities were detected.

Fe and Ag were deposited from e-gun and effusion sources , respectively, at a rate of ~0.5Å/s onto the substrate , held at room temperature. The substrate was rotated continuously at ~1rev/s during deposition. RHEED patterns recorded after Fe and Ag epitaxy were similar to those reported earlier [3]. In order to grow single orientation Ag films on GaAs it was necessary to first deposit at least 6ml of Fe. Subsequent Ag films grew with their (001) plane parallel to the (001) planes of Fe and GaAs(001) but with the Ag lattice rotated by 45° about the [001] axis. In this setting the lattice misfit between Fe and Ag is 0.8%.

Figure 1 shows a cross-section TEM micrograph of the interface between the GaAs and the Ag film. The interface is viewed along the [110] direction of the GaAs. The (200) planes of the Ag lattice are resolved. It is difficult to distinguish the 6ml Fe film from the Ag lattice but the dark band near the interface probably represents the Fe film. From this and other micrographs , in addition to in situ RHEED studies , it is clear that the Ag film is locked-in to a single orientation by the Fe film. For thinner or no initial predeposition of Fe we found that the Ag grew with a mixture of two different orientations: Ag(110) parallel to GaAs(001) or Ag(001) parallel to GaAs(001). This finding is in agreement with prior studies [5] of this interface. Details of our TEM studies of the Ag-Fe and GaAs-Fe interfaces will be presented elsewhere.

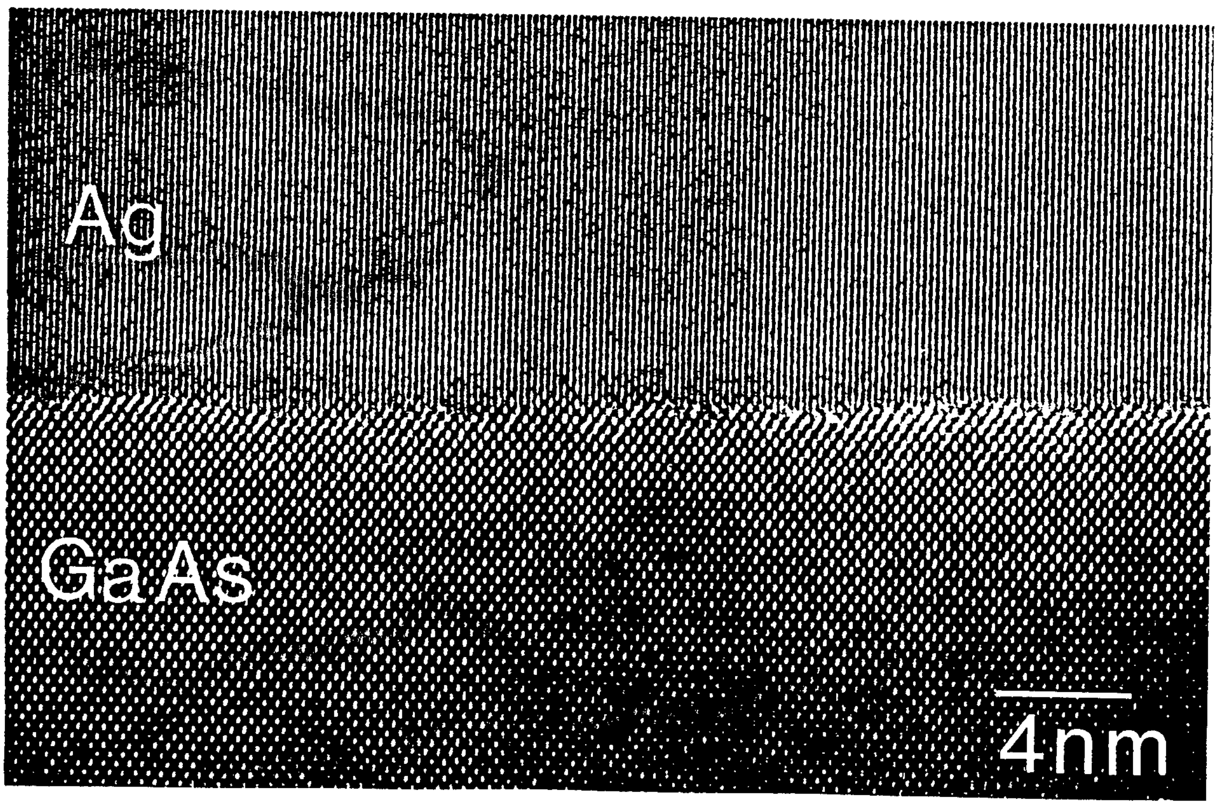

Figure 1 - Cross-section transmission electron micrograph of the interface region of a structure comprising: 500Å Ag / 6ml Fe / GaAs(001). The view is along GaAs[110]. The fringes in the Ag film represent the (200) planes.

DOUBLE CRYSTAL DIFFRACTOMETRY OF Fe-Ag-Fe-GaAs STRUCTURES.

Two types of structures were examined in this study. In the first (type 1) , a 500 or 2000Å film of Ag was grown onto the 6ml Fe film. This was followed by ~2000Å of Fe and a final protective film of 100Å Ag. In the second type (type 2) of structure a 2000Å film of Fe was grown onto the GaAs. This was followed by a 2000Å Ag film.

The sample crystal was probed by CuKα_1 radiation from the (004) reflection of a Si(001) monochromator. Both theta two-theta scans and rocking curves were recorded in symmetric and asymmetric settings [6]. Figure 2 shows theta two-theta scans for a type 1 structure comprising a 2000Å Fe film grown on a 2000Å Ag film. The scans were made in the symmetric setting for two different azimuths of the sample . As the sample was rotated about its surface normal the angular separations between Ag(002) and GaAs(004) peaks and between Fe(002) and GaAs(004) peaks varied cyclically with the cosine of the azimuthal angle. This is seen in Figure 2 where the Fe and GaAs peaks are nearly coincident for an azimuth of 12° but are separated by about 0.8 ° at an azimuth of 282° This behavior indicates that the [001] axes in the GaAs, Ag and Fe lattices are not coincident but point in different directions in space. We also found that the GaAs surface normal is not coincident with the GaAs [001] axis. Similar studies of the type 2 structures also revealed tilts of the various axes. Figure 3 illustrates the relative orientations of the axes in space for the type 1 and type 2 structures. The tilts are coplanar with the tilt of the substrate surface with respect to the GaAs(001) plane.

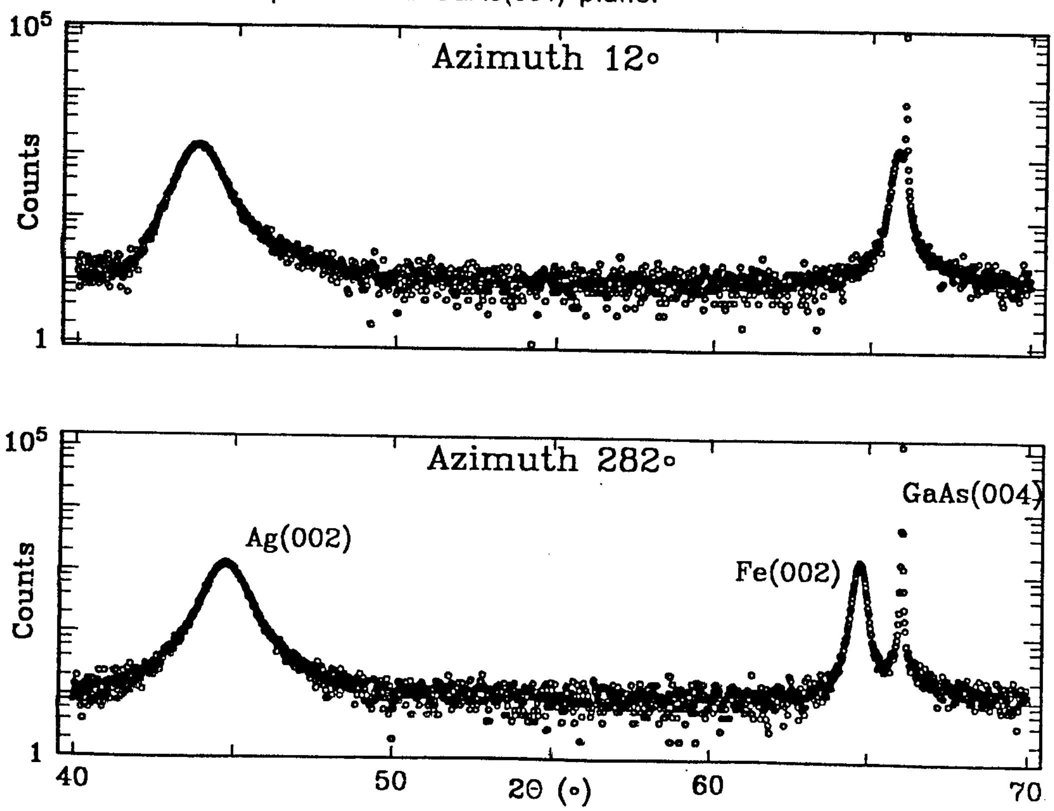

Figure 2 - Theta two-theta X-ray scans for epitaxial sandwich structure comprising : 100Å Ag / 2000Å Fe / 2000ÅAg / 6ml Fe/ GaAs(001). CuKα_1 radiation. The scans are for two different azimuths of the beam in the (001) plane of the GaAs substrate. Note that as the sample is rotated in its own plane , the Bragg peaks of Ag and Fe move relative to the GaAs(002) peak. This indicates tilts of the Ag[001] and Fe[001] axes away from GaAs[001].

Perpendicular strain ($\varepsilon^{\perp}$)values of the Fe films in type 1 structures were derived from the mean angular separation of Bragg peaks in rocking curves of the Fe(002) and GaAs(004) in the symmetric setting. Both the perpendicular and parallel strains ($\varepsilon^{||}$) were derived from the mean angular separation of Bragg peaks in rocking curves of Fe(022)and GaAs (044) in the asymmetric setting [6]. For symmetric reflections the angular separation between the Fe and and GaAs peaks is

$$\Delta\omega = -\varepsilon^{\perp}\tan\theta_B + \phi_m\cos\alpha \qquad 1$$

where θ_B is the substrate Bragg angle, ϕ_m is the tilt of the Fe [001] axis from the GaAs [001] axis and α is the azimuth.
For asymmetric reflections $\Delta\omega$ is related to both $\varepsilon^{\perp}$ and $\varepsilon^{||}$ by:

$$\Delta\omega = -(\varepsilon^{\perp}\cos^2\phi + \varepsilon^{||}\sin^2\phi)\tan\theta_B \pm (\varepsilon^{\perp} - \varepsilon^{||})\sin\phi\cos\phi + \phi_m\cos\alpha \qquad 2$$

where the $\pm$ sign refers to the two possible paths for the X-ray beam in in the asymmetric setting and ϕ is the angle between the Bragg planes and the sample surface. Tables 1 and 2 summarize the X-ray and elastic strains for Fe films derived from rocking curves for several type 1 structures and a type 2 structure. Strain measurements for the Ag films confirmed , that in both types of structures , the Ag films were strain-free and fully relaxed to the bulk lattice constant for Ag.

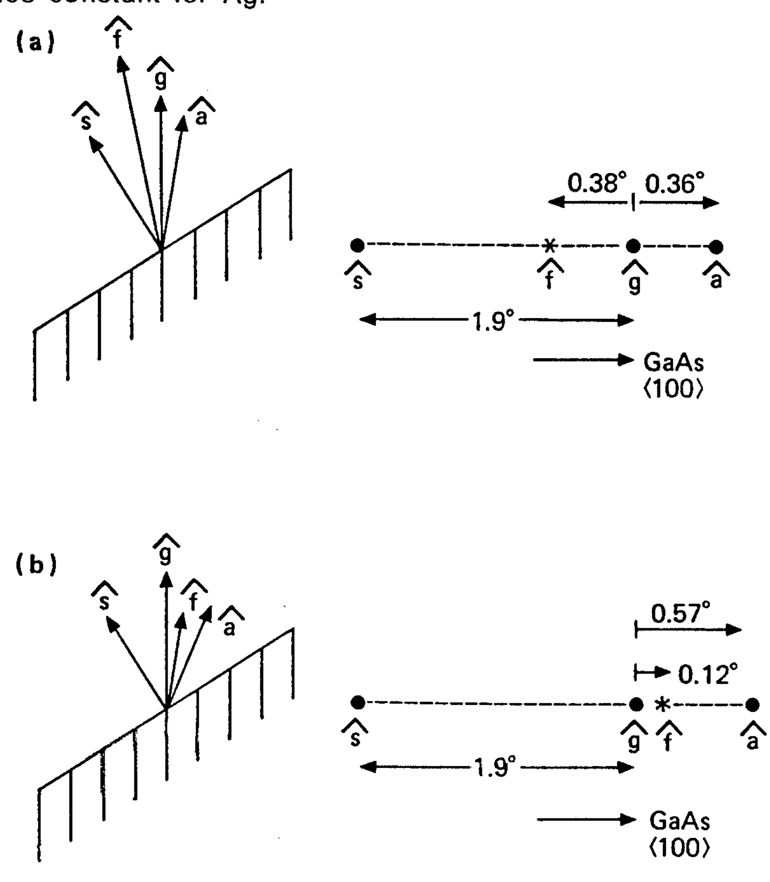

Figure 3 - Schematic diagram illustrating the directions of the axes in epitaxial Ag / Fe structures grown on GaAs(001).
a , f and g are the [001]axes of Ag , Fe and GaAs respectively. **s** is the GaAs surface normal.

a) Sample 138 : 100Å Ag / 2000Å Fe / 2000Å Ag / 6ml Fe / GaAs(001).
b) Sample 139 : 2000Å Ag / 2000Å Fe / GaAs(001).

TABLE 1

X-RAY STRAIN ($\varepsilon^{\perp}, \varepsilon^{||}$) AND ELASTIC STRAIN ($e^{\perp}, e^{||}$) FOR EPITAXIAL FILMS OF Fe IN Ag / Fe / Ag / GaAs(001) SANDWICH STRUCTURES.

SAMPLE	$\varepsilon^{\perp}$(%)	$\varepsilon^{||}$<100>	$\varepsilon^{||}$<010>	$\phi m^{(o)}$	$e^{\perp}$(%)	$e^{||}$(%)
MBE 138	1.02	1.69	1.70	0.38	-0.37	0.30
MBE 66	0.86	1.84	1.89	0.54	-0.52	0.45
MBE 67	0.86	1.81	1.81	0.60	-0.52	0.42

TABLE 2

X-RAY STRAIN ($\varepsilon^{\perp}, \varepsilon^{||}$) AND ELASTIC STRAIN ($e^{\perp}, e^{||}$) FOR AN EPITAXIAL FILM OF Fe IN Ag / Fe / GaAs(001) SANDWICH STRUCTURE.

SAMPLE	$\varepsilon^{\perp}$(%)	$\varepsilon^{||}$<100>	$\varepsilon^{||}$<010>	$\phi m^{(o)}$	$e^{\perp}$(%)	$e^{||}$(%)
MBE 139	1.63	1.14	1.13	0.12	0.24	-0.25

Notes:

1. Samples MBE 66 and 67 comprised 100Å Ag / 1875Å Fe / 500Å Ag / 6ml Fe / GaAs. Sample MBE 138 comprised 100Å Ag / 2000Å Fe / 2000Å Ag / 6ml Fe / GaAs. Sample MBE 139 comprised 2000Å Ag / 2000Å Fe / GaAs(001). All samples were grown and measured at room temperature (20°C).

2. $\varepsilon^{\perp}$ and $\varepsilon^{||}$ are the X-ray strains ($\Delta d/d$) between the doubled Fe unit cell and GaAs measured perpendicular and parallel to the (001) film plane.

3. $e^{\perp}$ and $e^{||}$ are the perpendicular and parallel elastic strains in the Fe film with respect to the relaxed bulk unit cell of Fe.

4. ϕ_m is the measured tilt of the Fe [001] axis with respect to the GaAs [001] axis. The tilt is along a <100> direction in the GaAs.

In the case of the asymmetric setting $\varepsilon^{||}$ and $\varepsilon^{\perp}$ were determined from two pairs of reflections : one pair with
$\alpha = 0$, beam incident at $\theta_B + \phi$; $\alpha = 180°$, beam incident at $\theta_B - \phi$
the other pair with
$\alpha = 90°$, beam incident at $\theta_B + \phi$; $\alpha = 270°$, beam incident at $\theta_B - \phi$
The former pair gave $\varepsilon^{||}$ along the <100> direction in GaAs , the second pair gave $\varepsilon^{||}$ along <010>. Tables 1 and 2 show that the in-plane strain is isotropic since the values of $\varepsilon^{||}$ are in good agreement (within 5%). The values of $\varepsilon^{\perp}$ for the symmetric and asymmetric settings were also in good agreement. Elastic strains for the Fe films were calculated from the X-ray strain values using room temperature (bulk) lattice constants for Fe and Ag of 2.8662Å and 4.08626Å respectively. These values are summarized in the last two columns of each table. Values of Poisson's ratio , derived from the data in tables 1 and 2 , were 0.39 and 0.32 respectively. The literature value [7] is 0.42.
In the case of type 1 structures the elastic strains show that the films are dilated in-plane and compressed along GaAs[001]. This is consistent with residual coherency strain arising from misfit between the Fe film and its Ag film substrate. The full values of coherency strain in this case would be $e^{||} = 0.8\%$ and $e^{\perp} = -1.16\%$. In the case of the type 2 structure the measured elastic strains indicate residual coherency strain arising from the misfit between Fe and the GaAs substrate. In this case the full value of coherency strain would be $\varepsilon^{||} = -1.39\%$ and $\varepsilon^{\perp} = 2.01\%$.

DISCUSSION.

The measured values of tilts in both type 1 and type 2 structures are qualitatively consistent with a recent model [8] for tilted epitaxy proposed by Bai et al. In this model the substrate surface is inclined at an angle ϕ_s to a low index plane . This breaks the surface symmetry and leads to a terraced growth surface with step edges along one direction. The total energy of the bicrystal system can then be lowered by a tilt of the film growth axis with respect to the surface normal. If the film lattice parameter in the growth direction is greater than the substrate parameter ($\varepsilon^{\perp} > 0$) then a tilt of the film axis away from the surface normal leads to matching of atoms in the plane of the interface along a direction normal to the step edges. Similarly, if $\varepsilon^{\perp} < 0$ then matching occurs when the film axis tilts towards the surface normal. The matching condition is satisfied when $\delta = \varepsilon^{\perp} \tan \phi_s$, where ϕ_s is the tilt of the substrate surface from the low index plane. In the present case the low index plane is (001) and the maximum tilt of the 6ml Fe prelayer should occur for a coherent interface for which $\varepsilon^{\perp} = 3.4\%$. The tilt would then be away from the surface normal by 0.065°. The maximum tilt for Ag on Fe is for a coherent interface for which $\varepsilon^{\perp} = 1.5\%$. In this case the Ag would tilt away from the surface normal by 0.029°. The total tilt of the Ag away from the GaAs surface normal would then be $\sim 0.065 + 0.029 = 0.094°$. We indeed measure a tilt of the Ag film away from the surface normal, coplanar with the tilt of the substrate surface , but by an amount (see Figure 3 a) much greater than this. Similarly , for the 2000Å Fe film on Ag we expect a maximum tilt for the case where the Fe is coherent with the Ag and $\varepsilon^{\perp} = -2.0\%$. In this case , Fe tilts towards the surface normal by 0.03°. Again we measure a tilt in this sense but an order of magnitude greater. A similar discrepancy occurs for the type 2 structures. Furthermore , since we observe misfit dislocations at each of the interfaces the predicted tilts are less than the values estimated above. At this stage the reason for the tilt enhancement

is not clear but we expect that full analysis of the high resolution images of the interfaces will suggest a mechanism.
The residual coherency strain of Fe films in both types of structure shows that strain relaxation to the thermal equilibrium value by nucleation and propagation of dislocations is inhibited by kinetics at room temperature. The complete relaxation of the Ag films may reflect the fact that Ag is mechanically softer than Fe and that dislocations may nucleate and propagate more readily. This phenomenon of residual coherency strain , at distances far from a lattice-mismatched interface , has also been observed [7] in the case of MBE-grown films of $CoSi_2$ on Si (111) surfaces. Such structures are metastable with respect to the nucleation of misfit dilocations.

CONCLUSIONS.

We find that epitaxial Fe films , grown at room temperature, on either GaAs or Ag film surfaces possess residual coherency strain at thicknesses of 2000Å The [001] directions of the Fe and Ag films are tilted with respect to the GaAs [001] axis. The directions of these tilts are in agreement with a recently proposed model for tilted epitaxy on misoriented substrates but the magnitudes of the measured tilts are much greater than predicted. Clearly , tilted epitaxy is a general phenomenon in growth on misoriented substrates and it must be included in any quantitative strain analysis of epitaxial structures.

ACKNOWLEDGMENTS

This work was partially supported by the office of Naval Research: Contract number N00014-87-C-0339.

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STRUCTURAL AND MAGNETIC CHARACTERIZATION OF RARE EARTH AND TRANSITION METAL FILMS GROWN ON EPITAXIAL BUFFER FILMS ON SEMICONDUCTOR SUBSTRATES.+

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+ This work was supported in part by ONR Contract N00014- 87-C-0339.

ABSTRACT

Structural and magnetic data are presented and discussed for epitaxial films of rare earth metals (Dy, Ho, Er) on LaF_3 films on the GaAs($\overline{111}$) surface and Fe on Ag films on the GaAs(001) surface. Both systems exhibit unusual structural characteristics which influence the magnetic properties of the metal films. In the case of rare earth epitaxy on LaF_3 we present evidence for epitaxy across an incommensurate or discommensurate interface. Coherency strain is not transmitted into the metal which behaves much like bulk crystals of the rare earths. In the case of Fe films, tilted epitaxy and long-range coherency strain are confirmed by X-ray diffractometry. Methods of controlling some of these structural effects by modifying the epitaxial structures are presented.

1.INTRODUCTION

In a recent paper [1] we described several new approaches to epitaxy of rare earth and transition metals. One aim of these approaches is to explore the effects of strain, imposed through coherency across epitaxial interfaces, on the magnetic properties of the films. In the case of the rare earth metals, basal plane epitaxy of Dy onto epitaxial films of LaF_3 grown onto the 3-fold symmetry GaAs($\overline{111}$) surface were described [1] In the present paper we describe the extension of this epitaxial system to Ho and Er films and report the characterization of the structures using double-crystal (DCD) and grazing incidence X-ray diffractometry (GID) in addition to XPS depth profiling and Rutherford Back Scattering Spectrometry (RBS). A major finding of these studies is that the rare earth lattice is rotated by 30° about the c-axis from the setting expected from in plane translational registry of the rare earth and LaF_3 lattices. Thus the interface is either incommensurate or discommensurate. Here we use the definition of interface type given by Gibson and Phillips [2]. In the former case there is no tendency for in-plane lattice matching of the overlayer to the substrate. In this case there are no interfacial misfit dislocations. On the other hand, the discommensurate interface is one in which the overlayer initially adjusts its in-plane lattice constant to match the substrate but subsequently relaxes to its normal lattice constant by formation of misfit dislocations. This is also known as a semi-coherent interface. At present we have insufficient experimental evidence to distinguish between the two cases. However, we do find that coherency strain is not transmitted across the interface and, as a result, epitaxial films of Dy, Ho and Er are strain-free over the thickness range (25-8000 Å). Although RBS shows no evidence for F contamination of the films, XPS depth profiling suggests that F is transported across the interface by as much as 500A in some cases. Thus it is possible that

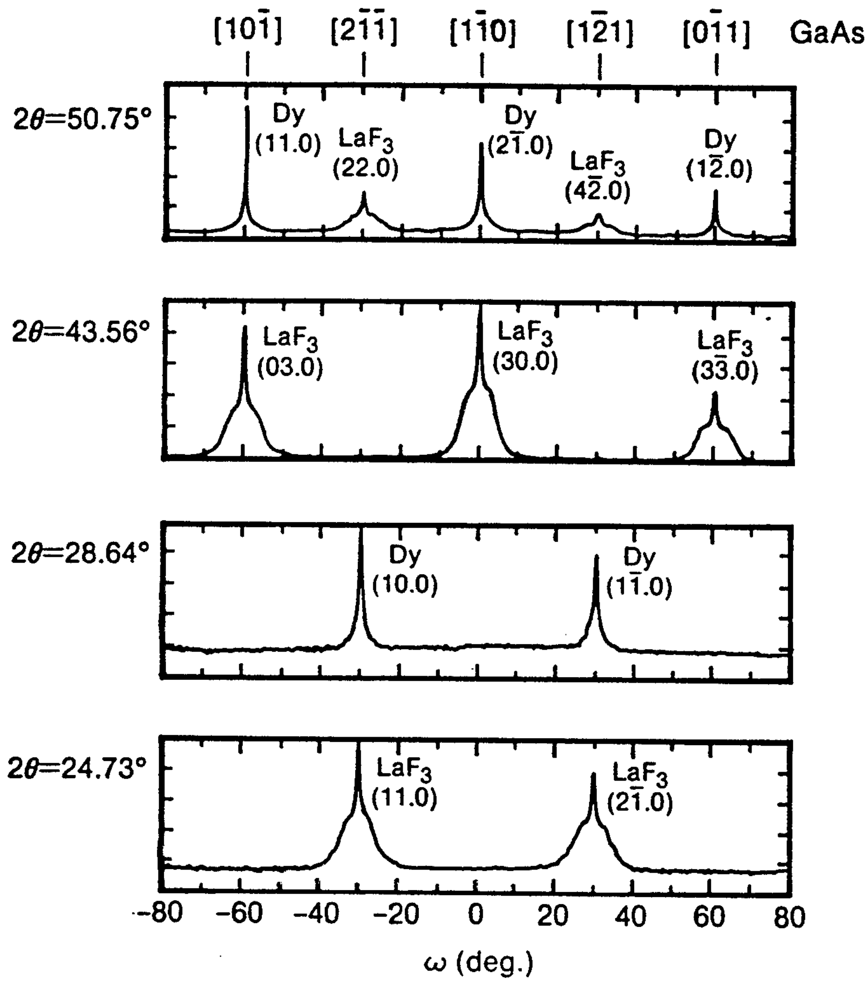


Figure 1 - GID ω scan for structure comprising
400ÅLaF_3 / 2000ÅDy/
2500ÅLaF_3 / GaAs($\bar{1}\bar{1}\bar{1}$).
Note the parallelism of Dy(10.0) and LaF_3(11.0)
planes indicating an orientational misfit between the two
lattices of 30°.

the magnetic properties of films thinner than this may be influenced by impurity F. These two findings prevented our original intention of exploring the magnetic implications of strain and dimensionality in rare earth films by adjusting coherency strain through lattice misfit across the rare earth - fluoride interface. However , by using a modified epitaxial structure we are still able to achieve this aim and at the same time eliminate the complicating effect of impurity outdiffusion of F.

In the case of epitaxy of Fe films on Ag films on GaAs (001) substrates we showed [1,8] previously that long range coherency strain and tilted epitaxy were present in Fe films grown on GaAs substrates cut 2° off the (001) plane. We find that tilted epitaxy is not present if growth occurs on exactly (within 0.12°) oriented substrates.

2. EXPERIMENTAL TECHNIQUES. The sample preparation techniques were identical to those described previously [1]. Double-crystal diffractometry .was carried out using an automated difractometer (manufactured by Blake Instruments) operating in the parallel setting with a Si (004) Bragg diffraction from the (001) face of a Si crystal. Slits were used to cut out the $CuK\alpha_2$ diffracted beam from the Si crystal. Grazing incidence diffractometry was carried out using the system described by Segmuller[3]. RBS studies of the films were carried out at the University of Arizona and at Almaden Research Center. SQUID magnetometry , and in some cases vibrating sample magnetometry was used to study the magnetic properties of the films.

3. EPITAXIAL LaF_3/Rare Earth Metal/ LaF_3 SANDWICH STRUCTURES.

As described previously for Dy [1,5] the rare earth metals Ho and Er were also found to grow epitaxially on LaF_3 films on GaAs($\overline{111}$)surfaces at 300°C. In all cases the LaF_3 growth was carried out at a substrate temperature of 500°C. This initial film was 1000-2000 Å thick. After growth of the rare earth film, a protective film of LaF_3 was grown at 300°C to protect the rare earth film from atmospheric oxidation. The sandwich structures were removed from the MBE system and characterized structurally and magnetically.

Figure 1 shows the GID data for a structure comprising 2500Å LaF_3 / 2000Å Dy/ 400Å LaF_3. The 4 scans are each for a particular 2θ value and the crystal is rotated about the GaAs(111) axis. The peaks correspond to Bragg diffraction from planes of Dy and LaF_3 which are parallel to the c-axis of these materials. The parallelism of Dy(10.0) and (11.0) LaF_3 planes is evident from the fact that the Bragg diffractions occur at the same crystal (ω) setting. The composite shape of the Bragg peaks for LaF_3 is due to superposition of peaks from the LaF_3 under and overlayers. The latter was grown at 300°C and is of lower structural perfection than the underlayer. Figure 2 shows schematically the relative orientation of basal planes of LaF_3 and Dy for (a) the setting in which Dy(11.0) is parallel to LaF_3(11.0) and (b) the observed setting . We had expected that the epitaxial relation would be as in (a) in view of the close(within 0.1%) match between the doubled (2a) lattice constant of Dy and the value of a for LaF_3 (7.1871Å in the space group $P6_3cm$. See reference 4). However , the observed relation has no translational registry between the lattices. The interface is therefore either incommensurate or discommensurate and in either case the lack of translational registry implies relaxation of the metal overlayer to its bulk lattice constant. Similar scans for sandwich structures of Ho and Er confirmed that these metals had identical epitaxial relations. Furthermore , in plane lattice constants

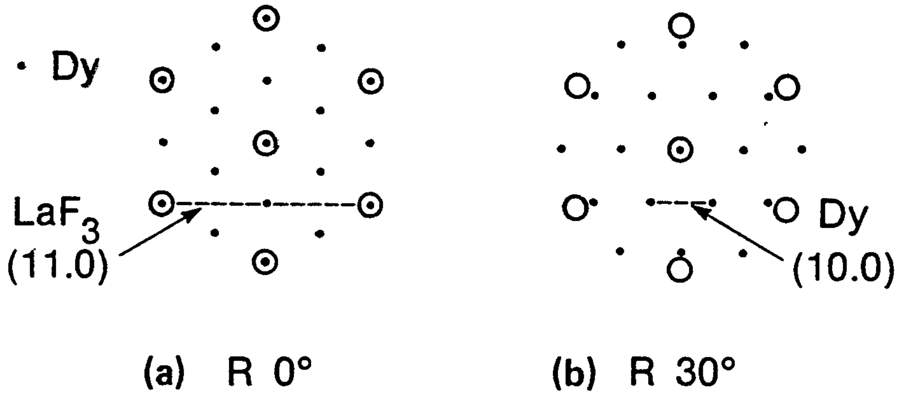


Figure 2 - Schematic diagram showing (a) the expected commensurate interface between LaF_3 (open circles) and rare earth lattices in the basal plane and (b) the observed 30° rotation. of Dy about the c-axis leading to an absence of translational registry between the lattices.

TABLE 1

In-Plane Lattice Constants and Coherency Lengths for Epitaxial Rare Earth Films, Determined by Grazing Incidence X-Ray Diffraction

	Thickness (Å)	a (Å)	l (Å)	Δa (%)	$a_{meas.}-a_{Bulk}$ (%)
Dy	2000	3.5945±0.001	180	0.03	+0.12
Er	4000	3.5586±0.0012	250	0.03	0.00
Er	100	3.5547±0.001	97	0.03	- 0.11
Er	50	3.5610±0.0014	80	0.04	+0.06
Ho	500	3.5773±0.0012	145	0.03	0.00
Ho	25	3.5780±0.001	65	0.03	+0.02

derived from ω and θ- 2θ scans showed that the metal lattice constant was indeed relaxed to the bulk value , even for films as thin as 25Å. Table 1 summarizes this data and the coherency length (l) for the films. The latter values were derived from the peak widths in the ω scans [3]. The column headed 'Δa' gives the standard deviation of the measured lattice constants. Clearly there is no significant coherency strain in any of the films. There is , however , a systematic trend towards smaller coherency lengths with decreasing film thickness. indicating a greater degree of in-plane disorder in the thinner films. XPS depth profiling suggested F transport across the interface by as much as 100Å for one structure containing a 500Å film of Dy grown from an effusion cell. The RBS studies showed no evidence for contamination of the films with F. However, the depth resolution and sensitivity of this technique is not as great as for XPS depth profiling. It seems likely therefore that the thinnest (<200Å) rare earth films may contain impurity F which introduces structural disorder leading to broadening of the magnetic transitions.

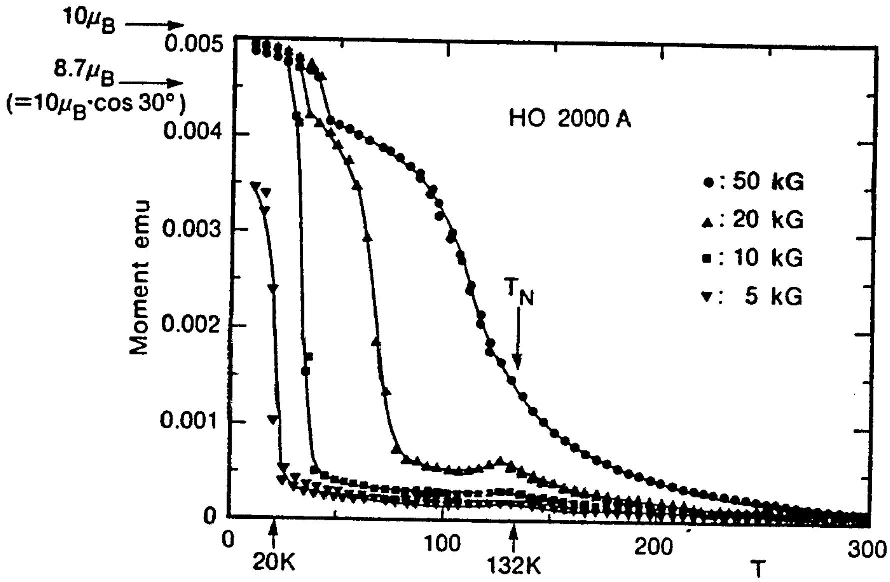
SQUID magnetometry data for two representative films of Ho are shown in Figure 3. The magnetic behaviour of the 2000Å film is identical with bulk Ho (see for example Coqblin[6]). In particular, the temperature of the transition to the helical state occurs at 132K but the ferromagnetic ordering is strongly field dependent. The field dependence of ferromagnetic ordering is similar to bulk. Note that the field is applied at 30° to the a-axis (the 'easy' direction). At the lowest temperatures the magnetisation approaches the bulk value of 10.4 Bohr magnetons per Ho atom. On the other hand , the data for the thin Ho film shows less abrupt transitions and the helical ordering transition is barely evident.

4. MODIFIED LaF_3 SANDWICH STRUCTURES: LaF_3/Er/Dy/Er/LaF_3.

In order to impose a uniform coherency strain on Dy it is necessary to establish a coherent interface on both sides of the Dy. We have arranged this by sandwiching the Dy between films of Er grown on the LaF_3. The Er film , as we have shown, has no strain imposed by the substrate. The Dy film , however, grows coherently on the Er and is subjected to coherency strain. The magnitude of the elastic strain in the Dy depends on the thickness of the Dy film and on the thickness of the Er under and overlayers. One would expect coherency strain to relax with Dy film thickness and that the maximum strain would occur for a thin Dy film sandwiched between much thicker Er films. In our test structure we sandwiched a 500Å film of Dy between a 2000Å underlayer and a 1000Å overlayer. The Dy and Er films were grown at 300°C. An added benefit of this structure was that F outdiffusion from the LaF_3 into the Er was limited to the near interface region. XPS depth profiling confirmed that the Dy film was free of impurity F.

Double crystal diffractometry studies [7] showed that the Dy film was indeed strained in the sense expected from the lattice misfit between Dy and Er. The perpendicular elastic strain was found to be $+0.27\pm0.02\%$ in the c-axis (ie. an expansion along the c-axis). On the other hand the Er film was close to the fully relaxed state with a perpendicular elastic strain of $-0.014\pm0.01\%$. Clearly, misfit dislocations have reduced but have not fully relaxed the coherency strain (maximum perpendicular value of 0.58%) in Dy.

Interestingly, SQUID measurements for this structure showed that the ferromagnetic ordering temperature for the Dy was higher than for bulk Dy. This is illustrated in Figure 4



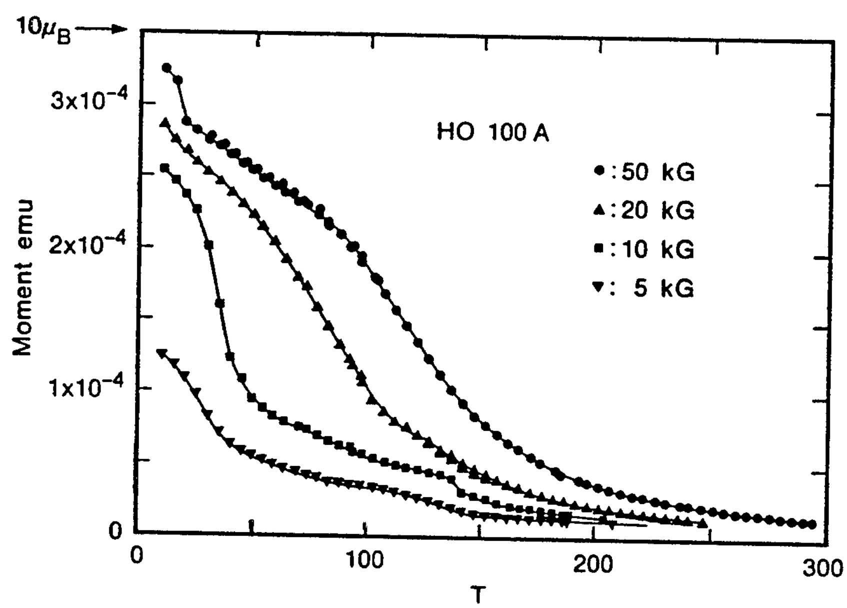


Figure 3 - SQUID magnetometer data for films of Ho in LaF_3 / Ho / LaF_3 sandwich structures. The magnetization is measured as a function of temperature for several values of applied field along the b direction in the basal plane.

(a) 2000Å Ho. (b) 100Å Ho.

Note that the saturation magnetization at low temperature and highest fields approaches the bulk value of 10 Bohr magnetons/ Ho atom.

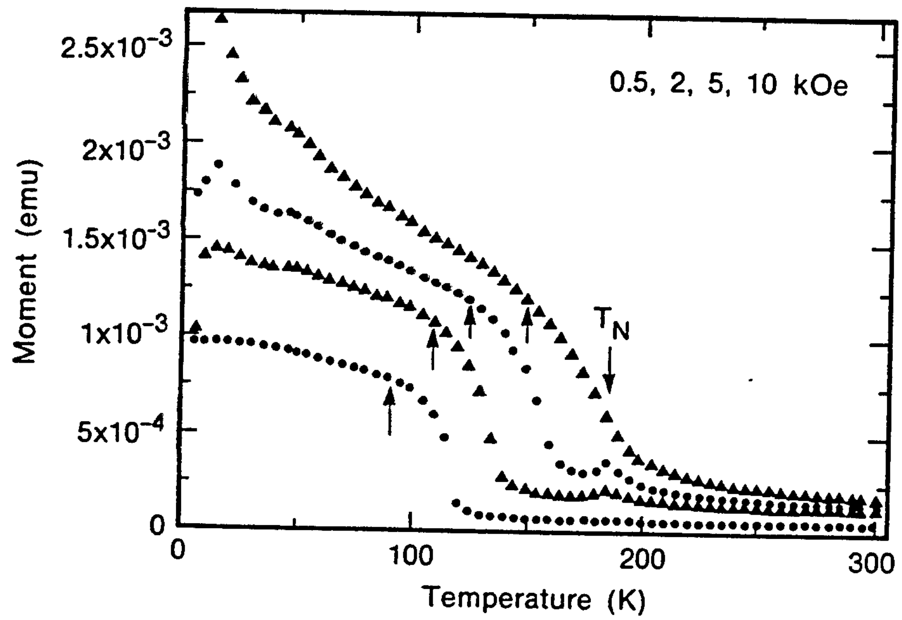


Figure 4- SQUID magnetometer data for a sandwich structure comprising 1000Å LaF_3/ 1000Å Er / 500Å Dy /2000Å Er / 50Å LaF_3 / GaAs(TTT).
Note the paramagnetic to helical transition at 178K and the helical-ferromagnetic transition at lower temperatures. The arrows indicate the onset of ferromagnetic ordering in bulk crystals. In each case the films order at higher temperature than bulk.

which shows data for 4 different values of applied field along the a-axis (easy axis of bulk crystal ie. [11.0]). The arrows indicate the onset of ferromagnetic ordering in bulk Dy. The ferromagnetic ordering in the film is in all cases completed at higher temperatures than the onset temperature for ordering in the bulk crystal. We attribute this to the imposed elastic strain in the film since, in bulk crystals, ferromagnetic ordering is accompanied by a discontinuous c-axis expansion and in-plane contraction. By mechanically imposing the structure of the ferromagnetic phase at a temperature above the bulk T_c we have made it energetically favourable for ferromagnetic ordering to occur at higher temperatures. The possiblility of interference from ferromagnetic ordering of the Er is not possible since ordering of the Er film occurs at much lower temperatures (<50K for fields below 10kOe) than Dy. In addition, differential thermal expansion between Er and Dy as the film is cooled from room temperature to 100K is less than 0.1% and would not significantly modify the much larger misfit-imposed strain in the Dy.

If a coherency strain of opposite sign were imposed on the Dy , by for example , growing it on an Y substrate then we would expect the ferromagnetic ordering to be suppressed. Data from Kwo et al.[10] does indeed show such an effect but no strain measurements were reported for those films.

5. EPITAXIAL Fe FILMS GROWN ON Ag FILMS ON GaAs(001).

In two recent papers [1,8] we reported long-range coherency strain and tilted epitaxy in Fe films grown on Ag films on GaAs(001) substrates. In that case the substrates were cut 2° off the (001) plane. We have recently found that tilted epitaxy does not occur when the GaAs(001) substrates are cut to within 0.12° of the (001) plane. Preliminary studies indicate that coherency strain is still present in Fe films grown on these substrates. It therefore seems likely that tilted epitaxy is not a strain relief mechanism as suggested by others[9], but is a result of the symmetry-breaking cut of the substrate. This view is supported by the in-plane isotropy [8] of elastic strain in the Fe films grown on the vicinal substrates. Clearly, tilt and coherency strain have different origins and are uncoupled.

6. DISCUSSION AND CONCLUSIONS.

The origin of the 30° rotation of the rare earth lattice on LaF_3 is presently unclear. However , one possibility is that the rare earth atoms in the overlayer bond preferentially to specific F atoms , in the LaF_3 surface , which occupy lattice sites rotated by 30° from the unit cell setting in Figure 2a. Alternatively , the observation of F in the near-interface region of the rare earth films may indicate a reactive interface , possibly with a second phase present. The GID data show no clear evidence for such a phase but simply indicate a greater degree of structural disorder in the thinnest (25Å) films. The lattice constant data also show no evidence of interstitial F in the rare earth films. High resolution TEM and in-situ photoelectron diffraction studies are planned to probe the interface.

The ability of the fluoride films to decouple the rare earth films from substrate-imposed strains is significant and distinguishes our structures from rare earth films grown by others; in particular from the data of Kwo et al [10] for Dy and of Flynn et al [11] for Er films on sapphire or Y substrates. In these latter cases, depression and suppression of ferromagnetic ordering is very clear and is attributed to 'lattice clamping' of the films to the substrate. Flynn et al. find that the magnetic properties of Er films remain strongly modified to film thicknesses as great as 8000Å. We on the other hand find that Er films grown on LaF_3 films on GaAs substrates are strain-free and behave similarly to bulk crystals at thicknesses greater than 500Å.

The prototype Er/Dy/Er sandwich structures define a promising approach to investigating the effects of strain on magnetic ordering in rare earths without the complicating artefact of F as an impurity. The preliminary result of an enhancement in T_c is interesting but needs to be confirmed by further samples with varying coherency strain in the Dy and by parallel studies of Dy/Y structures which should invert the strain in the Dy.

In the case of Ag/Fe/Ag/GaAs structures it remains to be seen to what extent the coherency strain and tilted epitaxy influence the perpendicular and in-plane anisotropy of the Fe films. Studies of these anisotropies in structures grown on exact orientation substrates are in progress. It is possible that the 2-fold in-plane anisotropy seen [12] in earlier structures grown on vicinal substrates may arise from the symmetry breaking effect of the substrate cut.

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Magnetic anisotropy and structural characterization of Co/Pt superlattices grown along selected orientations by molecular-beam epitaxy

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In this paper we report the molecular-beam epitaxial growth of Co/Pt superlattices on epitaxial Ag films on GaAs substrates. The growth axis of the superlattice was selected by seeded epitaxy to be along either the [001], [110], or [111] axis of Pt. The magnetic properties of the superlattices depend on the orientation of the growth axis. *In situ* x-ray photoelectron diffraction studies of the Co-Pt interfaces during their formation reveals that they are not atomically abrupt for any of the orientations. We conclude that models for magnetic anisotropy of Co/Pt superlattices should be based on a combination of magnetocrystalline anisotropy and strain in compositionally mixed interfaces.

I. INTRODUCTION

Artificially layered magnetic metal structures, including superlattices, are a topic of intense and current interest because of their unusual magnetic properties and for their potential in device applications. Over the past few years, these structures have demonstrated[1] a variety of novel phenomena such as perpendicular anisotropy, giant magnetoresistance, and magnetic exchange coupling. Numerous systems, such as Co-Au,[2] Co-Cu,[3] Fe-Ag,[4] Co-Pt,[5] and Co-Pd,[6] have already demonstrated that a magnetic easy axis can be obtained perpendicular to the interface planes when the thickness of the magnetic film is less than a few monolayers. However, the origin of this anisotropy remains unclear. Although there are indications that the magnetic and structural properties are closely related, it is often difficult to obtain a well-defined single-crystal structure. Molecular-beam epitaxy (MBE) provides a route to such structures since it is generally compatible with *in situ* structural characterization techniques during all stages of film growth. For example, using reflection high-energy electron diffraction (RHEED) with MBE growth techniques, den Broeder *et al.*[6] have demonstrated that the growth axis of Co/Pd superlattices strongly influences the magnetic anisotropy and conclude that coherency strain in Co is a key factor in this effect.

In this paper we report on the magnetic and structural behavior of MBE grown Co/Pt superlattices. Magnetic hysteresis loops are presented for three crystallographic orientations and compared with representative chemical and structural interface studies via x-ray photoelectron diffraction (XPD). The magnetic properties of the superlattices are reproducible and are highly dependent on orientation while the interfaces are chemically mixed to a limited extent. Significant strain in the [001] oriented Co overlayers is detected. Our results suggest that the magnetic anisotropy in Co/Pt superlattices arises from a combination of magnetocrystalline anisotropy and strain in compositionally mixed interfaces.

II. EXPERIMENTAL TECHNIQUES

The Co-Pt superlattices reported in this paper were prepared in a VG 80-M MBE system (VG Semicon Ltd.). The Co and Pt layers were grown from *e*-gun sources at rates of ~0.15 and ~0.25 Å/sec, respectively. The deposition rates and beam-interrupt shutters were computer controlled using a Sentinel III deposition control system (Leybold Heraus Inc.). Fluctuations in film thickness were less than 2%. Ag layers were grown at a rate of 0.4 Å/sec from a Knudsen cell held at ~1050 °C. The background pressure prior to film growth was approximately $\sim 2.10^{-11}$ mbar, while the pressure during superlattice growth was $\sim 2.10^{-10}$ mbar or better. The XPD studies were carried out in a modified VG Microlab II analysis chamber, which allowed for a ±4° solid angle to be scanned over the full 2π steradians hemisphere above the sample surface. All sample transfers from the growth chamber to the analysis chamber were carried out in fully UHV ($< 10^{-10}$ mbar) conditions. All measurements used AlK_α x rays as the excitation source.

III. SUPERLATTICE GROWTH AND STRUCTURE

The key to successful Co-Pt growth in the three different orientations hinged on the initial epitaxy of the Ag buffer layer which seeds parallel epitaxy of the isostructural Pt in each orientation and thus governs the orientation of the entire superlattice. Figure 1 shows a schematic diagram of the selected-orientation epitaxial superlattices. Details of the growth techniques are described elsewhere.[7]

IV. MAGNETIC MEASUREMENTS

Room-temperature magnetic measurements were made on the superlattices using the polar Kerr effect (MOKE, 633 nm), vibrating sample magnetometry, and torque magnetometry. Figure 2 shows the results of the MOKE measurements for three different orientations, 15-period superlattices, each of the composition ≃3 Å Co–16.7 Å Pt. The [111] sample exhibits a highly square loop with a coercivity of 3.2 kOe. This sample has remanent

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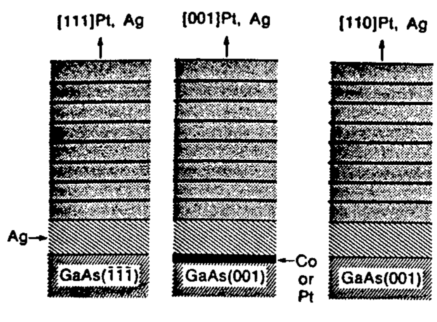

FIG. 1. Schematic diagram of the selected-orientation epitaxial Co/Pt superlattices. In each case the superlattices were grown on a Ag film, but the orientation of this film was selected by different growth procedures.

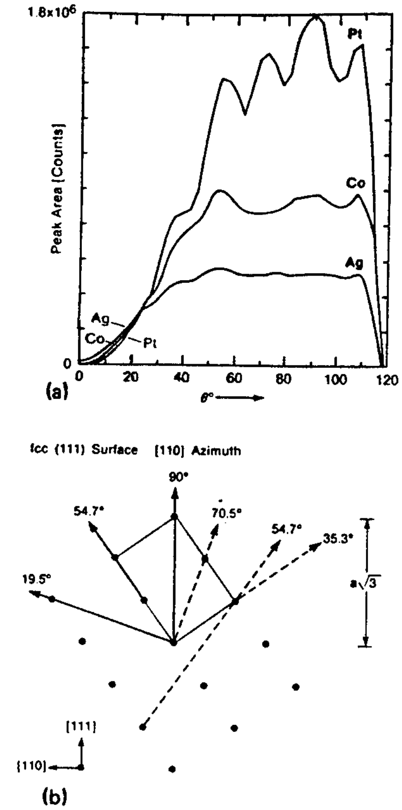

FIG. 3. (a) Polar x-ray photoelectron diffraction scan from a 3-Å Co film on a 17-Å Pt film recorded during the growth of a [111] oriented Co 3 Å–Pt 17 Å superlattice. Scan plane (110). (b) Schematic diagram of near-neighbor directions for forward scattering in a fcc lattice. (111) surface; [110] azimuth.

magnetization perpendicular to the film plane. On the other hand, the [001] sample has no remanence and cannot be saturated for the highest field (16 kOe) used, clearly suggesting a strong in-plane anisotropy. The [110] sample has intermediate behavior with both in-plane and out-of-plane remanence.

V. *IN SITU* STUDIES OF INTERFACE FORMATION BY X-RAY PHOTOELECTRON DIFFRACTION

The formation of Co-Pt interfaces was studied using angle-resolved x-ray photoelectron spectroscopy to probe forward scattering of photoelectrons from both the Co and the Pt films, as well as the Ag buffer film, when used. It has been known since the work of Fadley and Bergström[8] on single-crystal gold substrates, that the emission intensity of photoelectrons is strongly anisotropic due to the underlying crystal structure. In its simplest form, the angular distribution of forward scattered photoelectrons will have intensity maxima corresponding to near-neighbor atomic positions in the sample. The structure and composition of an interface can then be determined by measuring the polar and azimuthal angular distributions as a function of overlayer coverage during the formation of the interface. This approach has already proven to be very useful in several metal on metal interface studies.[9–12]

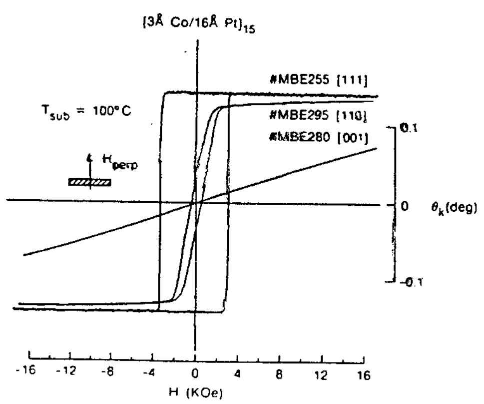

FIG. 2. Magnetic hysteresis loops recorded at 20 °C by Kerr rotation for oriented Co/Pt superlattices with ($\simeq 3$ Å Co–16.7 Å Pt)×15 periods. Wavelength of incident laser 0.633 nm. The magnetic field was applied normal to the sample surface, i.e., along the directions indicated.

Polar and azimuthal scans were obtained for both the [111] and the [100] oriented films using the Co $2p_{3/2}$, Pt $4d_{5/2}$, and Ag $3d_{5/2}$ core levels for Co coverages of 1, 2, and 3 Å. A study was also made of a 3-Å Co film deposited on a thick ($\sim$200 Å) Pt film, without a Ag buffer layer, for comparison. The angular scans were supplemented by angle-averaged survey scans, done with various depth sensitivities to determine the composition at the interface. Figure 3(a) shows a polar scan for a 3-Å Co film on a 17-Å Pt film recorded during the growth of a [111] oriented superlattice. Several features are clear from the data. Firstly, Ag is present in the probed region. Moreover, at low ($\lesssim 20°$) angles, that is, at more surface sensitive geometries, the Ag intensity becomes larger than the Pt or Co. This crossover behavior indicates segregation of Ag to the surface where the estimated coverage of Ag is approximately 1.5 monolayers (ML). (After 5 Co/Pt bilayers were grown on top of the Ag, the coverage was reduced to $\sim$0.3 ML.) Secondly, diffraction peaks at $\theta = 90°$, 72°, 56°, and 36° are present for Pt, indicating an fcc lattice. Even though the Co shows the initial stages of fcc symmetry, the peak at 72° is absent and the relative intensity of the 90° peak is weaker than that for Pt indicating reduced Co coordination. The Ag scan shows very weak diffraction features at 56° and 36° arising in part from the buried Ag seed film. The presence or absence of fine details in diffraction patterns can be complicated, but the gross features can usually be attributed to the atomic geometry about the emitter. In the case of Pt, which is 17 Å = 7.5 ML thick, one expects all forward scattering peaks indicated in Fig. 3(b) to be present, and they clearly are. The forward scattering peaks at 90°, 70.5°, 54.7°, and 35.3° require a minimum of 4, 3, 2, and 2 Pt layers, respectively. However, for Co, the presence of a

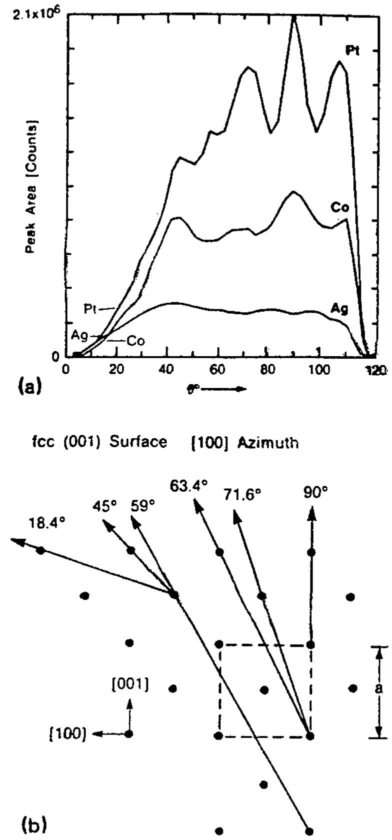

FIG. 4. (a) Polar x-ray photoelectron diffraction scan from a 3-Å Co film on a 17-Å Pt film recorded during the growth of a [001] oriented Co 3 Å–Pt 17 Å superlattice. Scan plane (100). (b) Schematic diagram of near-neighbor directions for forward scattering in a fcc lattice. (001) surface; [100] azimuth.

weak 90° peak suggests that the Co atoms are buried to at least 3 ML. Since the amount of Co deposited corresponds to only 3 Å = 1.5 ML, and the Co to Pt ratio remains constant over the low take-off angle region, we conclude that the Co film has mixed with Pt. In the case of the [001] oriented multilayer the polar scan shown in Fig. 4(a) again reveals surface-segregated Ag corresponding to a coverage of approximately 0.5 ML, which is a factor of 3 less than that for the (111) surface. (Crystal twinning present in the [111] oriented films, as seen from LEED and azimuthal XPD scans, may enhance Ag diffusion through added grain boundaries.) Diffraction peaks at 90°, 72°, 58°, and 45° are present for Pt as predicted in Fig. 4(b). These correspond to 3, 4, 6, and 2 layers of atoms respectively. Here again, with less than 2 ML of Co deposited, clustering and Co-Pt mixing can be inferred from the forward scattering peaks present. The Co peak at 90° is strong, suggesting a well-defined three-layer system. The 72° peak is much weaker and the 58° peak absent, defining the upper thickness limit to be less than 6 layers.

A 3-Å Co film was also deposited on a thick Pt [001] film without using the Ag seed layer. These results, although not shown, are in full agreement with the general results discussed thus far. It is interesting to note that although this system shows reduced long-range order, as seen by RHEED and LEED, the short-range order as seen by XPD has increased. The magnetic data showed no significant difference.

Lattice strain in the [001] oriented Co films is evident from slight shifts in diffraction peak positions relative to the Pt peaks. For example, along the [101] direction, diffraction occurs at 45° for Pt but is shifted to 43° for Co. This is consistent with a tetragonal distortion of the Co environment which has a reduced c/a (perpendicular to in-plane lattice constant) ratio of 0.9 ± 0.04. Such a distortion may be a result of coherency strain. On the other hand, the [111] oriented films exhibit no significant distortion. Detailed studies of this structural distortion are still in progress and will be reported subsequently.

VI. CONCLUSIONS

We have observed a strong orientation dependence of the magnetic hysteresis loops in Co/Pt multilayers prepared by MBE. Detailed analysis on representative films by XPD have shown the Co and the Pt to be mixed, retaining epitaxy over a 2–4 layer region with a slight cubic-to-tetragonal distortion in the case of the [001] oriented Co films. Ag, used as a seed layer for epitaxy, was found to surface segregate readily through the films, but it was determined that it did not influence the magnetic data through analysis of a control structure free of Ag. We conclude that models for the magnetic anisotropy of Co/Pt superlattices should be based on magnetocrystalline anisotropy of compositionally mixed interfaces with contributions from strain and possibly local alloy ordering.

ACKNOWLEDGMENT

This work was supported in part by the Office of Naval Research.

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Appendix 2.

Year end reports/summaries.

PUBLICATIONS / PATENTS / PRESENTATIONS / HONORS REPORT

for

1 October 1987 through 30 September 1988

for

Contract NOOO14-87-C-0339

R&T No. 414e045

INVESTIGATION OF THE EPITAXIAL GROWTH AND CHARACTERIZATION OF HIGH PERFECTION MAGNETIC THIN FILMS.

Principal Investigators: Robin F.C. Farrow and Stuart S.P. Parkin

IBM Almaden Research Laboratory

650 Harry Road, San Jose, CA 95130-6099

Reproduction in whole, or in part, is permitted for any purpose of the United States Government.

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A. Papers Submitted to Refereed Journals.

1. R.F.C. Farrow, S.S.P. Parkin, V.S. Speriosu, C. Chien, J.C. Bravman, R.F. Marks, P.D. Kirchner, G.A. Prinz, B.T. Jonker " Long-Range Coherency Strain and Tilted Epitaxy in Ag-Fe-Ag Sandwich Structures on GaAs(001)". Mat. Res. Soc. Symp. Proc., 102 (1988)

B. Papers Published in Refereed Journals

1. R.F.C. Farrow , S.S.P. Parkin, V.S. Speriosu, " New Approaches to Epitaxy of Transition Metals and Rare Earths: Heteroepitaxy on Lattice-Matched Buffer Films on Semiconductors." J. Applied . Phys. 64 (10), 15 November 1988. p. 5315.

2. " MBE Growth and Properties of Fe Films on Lattice-Matched InGaAs Films". by R.F.C. Farrow, S.S.P. Parkin, R.B. Beyers, M. Lang, V. Speriosu, P. Pitner, J.M. Woodall, S.L. Wright, P.D. Kirchner, G.D. Pettit., Mat. Res. Soc. Symp. Proc.102, 483, 1988.

3. " Epitaxial Growth of Rare Earths on Rare Earth Fluorides and Rare Earth Fluorides on Rare Earths : Two New Epitaxial Systems Accessed by MBE". by R.F.C. Farrow, S.S.P. Parkin, M. Lang, K.P. Roche, Mat. Res. Soc. Symp. Proc. 103, 205 ,1988.

C. Miscellaneous Publications

D. Invited Presentations

" A New Family of Epitaxial Systems for Probing the Effects of Elastic Strain on the Magnetic Ordering of Rare Earth Metals".(Invited). R.F.C. Farrow , presentation to John Armstrong, Director of IBM Research , September 14 , 1988 ,

"Magnetic Properties of Epitaxially-Grown Rare Earth Films." (Invited). S.S.P. Parkin, R.F.C..Farrow , International Conference on Magnetism, Paris, France , July 25-29 , 1988.

" New Approaches to Epitaxy of Rare Earth and Transition Metals: Heteroepitaxy on Lattice-Matched Buffer Layers on Semiconductors." (Invited) R.F.C. Farrow, S.S.P.Parkin , V.S.S. Speriosu. 4th Joint Magnetism and Magnetic Materials-Intermag Conference , July 12-15 , 1988 . Vancouver , Canada.

E. Contributed Presentations

"Long Range Coherency Strain and Tilted Epitaxy in Epitaxial Ag-Fe-Ag Sandwich Structures on GaAs(001) Substrates." R.F.C. Farrow, C.A. Shen, V.S.Speriosu, S.S.P. Parkin, C.J. Chien, J.C. Bravman, R.F. Marks, P.D. Kirchner. Materials Research Society Fall Meeting, Boston, Mass., November 28-December 3, 1988.

R.F.C. Farrow , S.S.P. Parkin , R.B. Beyers , M. Lang , V. S. Speriosu , P. Pitner , J.M. Woodall , P.D. Kirchner , G.D. Pettit, " MBE Growth and Interfacial

Properties of Fe Films on Lattice-Matched InGaAs Films". Fifteenth Annual Conference on The Physics and Chemistry of Semiconductor Interfaces, Asilomar, Ca February 1-4 , 1988.

F. Honors/Awards/Prizes

G. Postdoctorals Supported for the Year Ending 1 October 1988

1. Dr. Alex Bezinge , ETH Zurich. Febuary 1987 through August 1988.

H. Professional Activities

1. R.F.C. Farrow, Member of Advisory Panel : DARPA -URI on Diluted Magnetic Semiconductors.

2. R.F.C. Farrow, Meeting Chair : Materials Research Society 1989 Spring Meeting.

December 8 , 1988.

Dr. George Wright
Naval Research Laboratory
Washington DC20375

Dear George

With reference to our ONR contract " Investigation of The Epitaxial Growth of High-Perfection Magnetic Thin Films" (N 00014-87-C-0339) a description of the major achievements of our second year follows.

1. Using our newly discovered epitaxial system of basal plane epitaxy of rare earth metals on rare earth fluorides we have prepared strain-free films of Dy, Ho, and Er. We find no suppression of the Curie point in these films down to 25 Å in thickness and attribute this to the absence of coherency strain. We do find that the antiferromagnetic ordering transition is suppressed below about 100Å and attribute this to a dimensionality effect.

2. We have carried out the first full structural analysis of epitaxial Ag-Fe-Ag sandwich structures grown on GaAs. This was done using XTEM and double crystal diffractometry. The interfaces are semi-coherent and we find residual coherency strain in the Fe films even at thicknesses of 2000Å Clearly , misfit dislocations are not mobile in the Fe at room temperature. Tetragonal distortion of the Fe unit cell is confirmed.

All best wishes ,

Yours sincerely ,

Robin

October 10, 1990.

Dr. George Wright
Code 613
Office of Naval Research
800 N Quincy Street
Washington DC20375

Dear George

Please find below a brief summary of the highlights of work on our contract N00014-87-C-0339 for the period October 1989 - October 1990.

MBE GROWTH AND SPIN-POLARIZED PHOTOELECTRON DIFFRACTION STUDIES OF MAGNETIC FILMS AND SURFACES.

B.D. Hermsmeier, R.F.C. Farrow, S.S.P. Parkin. N00014-87-C-0339.

Considerable progress has been made this year in fulfilling our commitment to ONR. An automated X-ray photoelectron diffraction (XPD) system is now complete and is fully compatible with our MBE system, giving us the unique ability to make in situ structural as well as short range magnetic order measurements on MBE-grown films. Insight into the growth mode and development of epitaxial interfaces has been obtained in several systems including Co/Pt, Co/Ag, NdF_3/LaF_3, Mn/GaAs, and Pt/GaAs. Lattice symmetry relations between the substrate and overlayer or between successive overlayers can be established directly. Misfit-induced strain can also be measured, as we have shown in the case of Co/Pt multilayer structures. Polar and azimuthal scans revealed a decrease in the *c/a ratio* for a 3Å Co film grown on Pt(001). Interfacial mixing was also evident and an upper limit established. In addition, we have shown that XPD polar scans can provide detailed concentration profiles for the top 30Å of a structure ; a

thickness regime in which uncertainty in interface structure and composition often exists.

Three spin-polarized photoelectron diffraction (SPPD) experiments have been initiated. These are :

(1) Studies of short range magnetic order in the surface of the antiferromagnetic insulator MnF_2. Initial results suggest a new, high-temperature, short-range magnetic ordering transition at 380K which is 313K above above the Néel temperature.

(2) Synchrotron studies have begun, in collaboration with J. Lecante's group at L.U.R.E. in Orsay, France, on MnF_2 to further investigate this new ordering transition and its dependence on surface orientation.

(3) Studies of magnetic ordering behavior in localized Mn and Fe clusters grown by MBE on GaAs(110) substrates. SPPD is uniquely suited for this type of study since it is truly a short range order probe studying a short range effect.
All best wishes and looking forward to seeing you in San Diego.

Yours sincerely ,

Robin

September 26, 1991

Dr. George B. Wright
Office of Naval Research
Physics Division
Code 111455
800 N Quincy Street
Arlington
Virginia 222217

Dear George:

Please find below a brief summary of the highlights of work on our contract N00014-87-C-0339 for the period October 1990 - October 1991.

MBE GROWTH AND SPIN-POLARIZED PHOTOELECTRON DIFFRACTION STUDIES
OF
MAGNETIC FILMS AND SURFACES.

R.F.C. Farrow, B.D. Hermsmeier, S.S.P. Parkin. N00014-87-C-0339.

We have recently begun a new series of experiments to further our efforts in understanding the magnetic behavior of constructed films. The technique, magnetic circular x-ray dichroism is an external probe that gives element specific magnetization measurements. In collaboration with J. Stohr, M. Samant, Y. Wu, and D. Weller, other IBM employees, we have investigated Co/Pt multilayers prepared by sputtering, MBE and e-beam evaporation on the Lawrence livermore / SSRL beam line. (IBM is in the process of commissioning its own beam line at SSRL which will help facilitate related experiments in the future.) These studies reveal the presence of cobalt oxide in the sputtered and e-beam evaporated samples but not in the MBE samples. This observation may be related to our finding that the perpendicular anisotropy energy for {111} oriented multilayers is highest for the MBE samples, all other parameters remaining equal.